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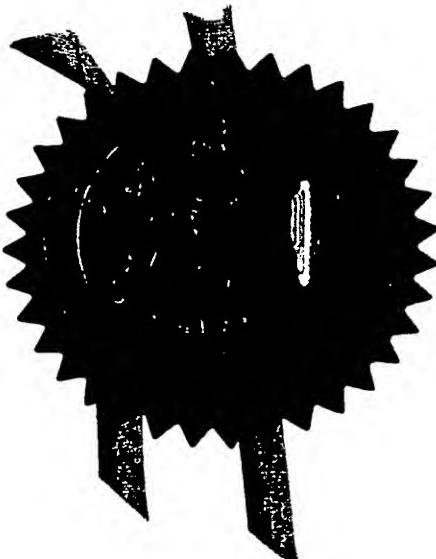
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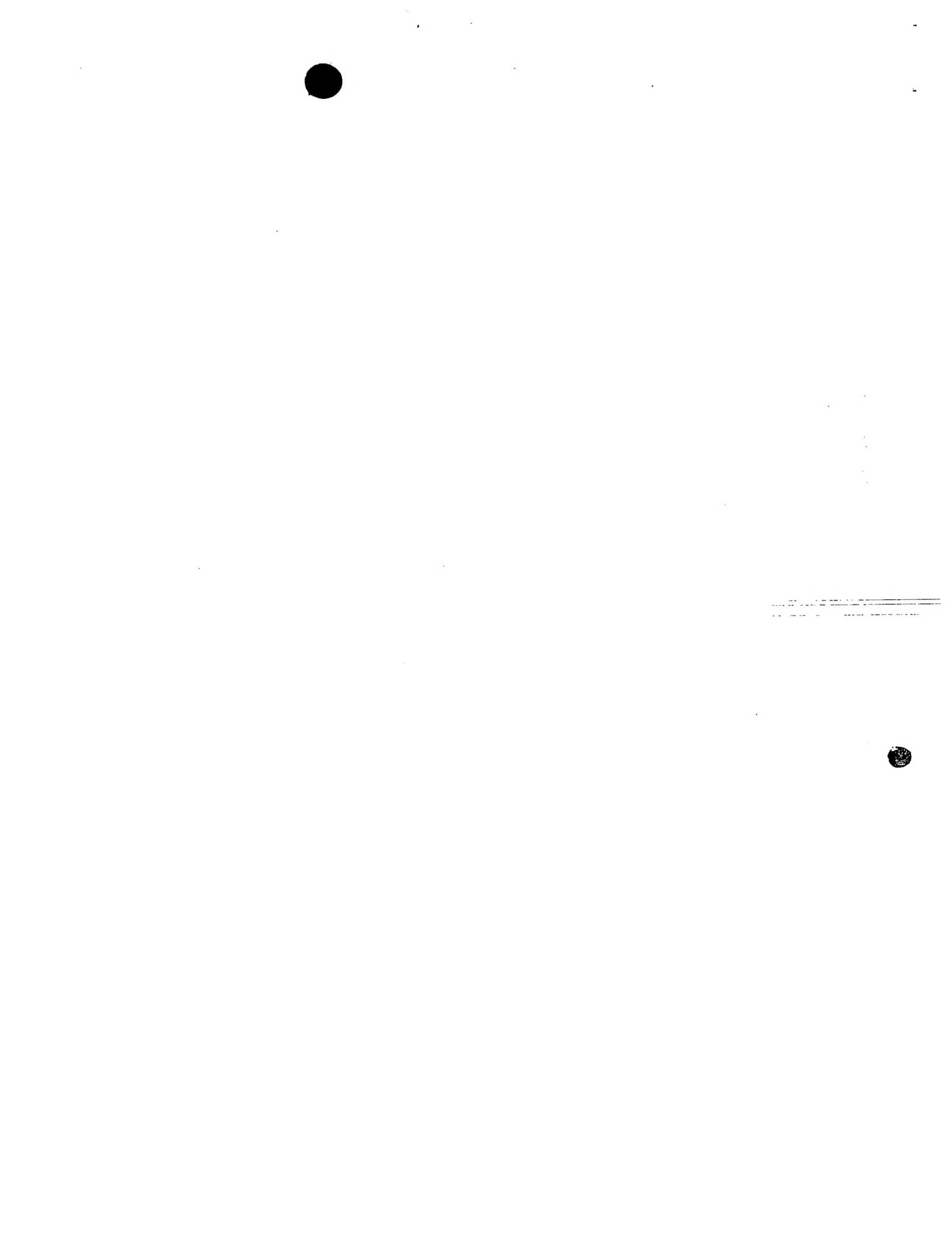


Signed

R. Mahoney

Dated

30th June 2000



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## Request for grant of a patent

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1/77

01DEC99 E495920-1 D00011  
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1. Your reference

SJA/OWK/53278/000

30 NOV 1999

2. Patent application number

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9928344.2

3. Full name, address and postcode of the or of each applicant (*underline all surnames*)NATIONAL POWER PLC  
Windmill Hill Business Park  
Whitehill Way  
Swindon  
Wiltshire  
SN5 6PBPatents ADP number (*if you know it*)

If the applicant is a corporate body, give the country/state of its incorporation

UNITED KINGDOM

5611011002

4. Title of the invention

ELECTROLYTE REBALANCING SYSTEM

5. Name of your agent (*if you have one*)BOULT WADE TENNANT  
27 FURNIVAL STREET  
LONDON  
EC4A 1PQ"Address for service" in the United Kingdom to which all correspondence should be sent (*including the postcode*)

42001

Patents ADP number (*if you know it*)If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (*if you know it*) the or each application number

Country

Priority application number  
(*if you know it*)Date of filing  
(*day/month/year*)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing  
(*day / month / year*)8. Is a statement of inventorship and of right to grant of a patent required in support of this request?  
(Answer 'Yes' if:

- a) any applicant named in part 3 is not an inventor, or
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Description 16 *(Handwritten mark)*

Claim(s) 4

Abstract

Drawing(s) 8 *(Handwritten mark)*

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Priority documents

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Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

Request for substantive examination  
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I/We request the grant of a patent on the basis of this application.

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*Susan Allard*

30 November 1999

12. Name and daytime telephone number of person to contact in the United Kingdom SUSAN J. ALLARD  
020 7430 7500

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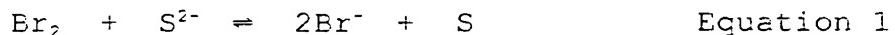
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### Electrolyte Rebalancing System

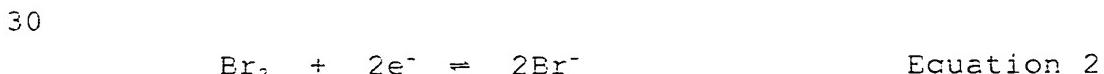
The present invention relates to the field of regenerative fuel cell (RFC) technology. In 5 particular it relates to apparatus and methods for the operation of RFCs which enhance their performance characteristics.

The manner in which RFCs are able to store and deliver 10 electricity is well known to those skilled in the art. An example of an RFC is described in US-A-4485154 which discloses an electrically chargeable, anionically active, reduction-oxidation system using a sulfide/polysulfide reaction in one half of the cell 15 and an iodine/iodide, chlorine/chloride or bromine/bromide reaction in the other half of the cell. The two halves of the cell are separated by a cation exchange membrane.

20 The overall chemical reaction involved, for example, for the bromine/bromide-sulfide/polysulfide system is shown in Equation 1 below:



25 However, within an RFC such as that described in US-A-4485154, the reaction takes place in separate but dependent bromine and sulfur half-cell reactions as shown below in Equations 2 and 3:



35 The sulfur produced in Equations 1 and 3 forms soluble polysulfide species in the presence of sulfide ions.

When the RFC is discharging, bromine is converted to bromide on the +ve side of the membrane and sulfide is converted to polysulfide on the -ve side of the membrane. Equation 1 goes from left to right and metal ions flow from the -ve side of the membrane to the +ve side of the membrane to complete the circuit. When the RFC is charging, bromide is converted to bromine on the +ve side of the membrane and polysulfide is converted to sulfide on the -ve side of the membrane.

5                  Equation 1 goes from right to left and metal ions flow from the +ve side of the membrane to the -ve side of the membrane to complete the circuit.

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The discharge/charge cycle described above will be repeated many times during the lifetime of the RFC and in order for the RFC to work efficiently throughout its lifetime it is important that the electrolytes remain balanced. In the context of the present specification, when the term "balanced" is used to describe the electrolytes it means that the concentrations of the reactive species within the electrolytes are such that both half-cell reactions are able to progress substantially to completion without one reaching completion before the other.

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Similarly, in the context of the present specification, the term "rebalancing" refers to a process which alters the concentration of one or more reactive species in one or both of the electrolytes so as to return said electrolytes to a balanced state or so as to maintain said electrolytes in a balanced state.

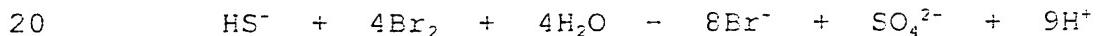
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At the beginning of the RFC's lifetime the concentrations of the reactive species on either side of the membrane will normally be fixed so that the electrolytes are balanced. However, once the RFC begins to operate in its repeating discharge-charge

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cycle, factors may intervene which result in the electrolytes becoming unbalanced. These factors will vary depending upon the identity of the reactive species within the electrolytes and on the manner in  
5 which the RFC is constructed and operated.

In the case of the bromine/bromide-sulfide/polysulfide RFC such as that described above, the most important factor which results in the electrolytes becoming unbalanced is the diffusion of unwanted species across  
10 the membrane. Although a cation selective ion-exchange membrane is used, during extended cycling of the cell some anionic species diffuse through the membrane. Thus, sulfide ions diffuse from the  
15 sulfide/polysulfide electrolyte into the bromine/bromide electrolyte where they will be oxidised by the bromine to form sulfate ions as shown in equation 4 below:



Equation 4

Other features which could similarly contribute to the  
25 above process are ineffective sealing between cell compartments, or catastrophic failure of any of the cell separating components, each of which may result in crossover of the electrolytes between cell compartments.

30            The oxidation of the sulfide goes beyond that which occurs during normal operation of the RFC. That is to say, the sulfide ions are oxidised all the way to sulfate ions and consequently consume four bromine  
35 molecules per sulfide ion rather than the normal one bromine molecule per sulfide ion which is consumed in the reaction scheme of Equation 1. As a result, the

bromine/bromide electrolyte becomes discharged to a greater extent than the sulfide/polysulfide electrolyte. Thus, when the cell is discharging there is insufficient bromine present to complete the discharge cycle. As a result, the voltage generated by the cell begins to decline earlier in the discharge cycle than when the electrolytes are balanced.

It would be advantageous to provide a process for rebalancing the electrolytes in order to compensate for the unbalancing effect of the crossover of sulfide and/or polysulfide electrolyte into the bromine electrolyte. Accordingly, the present invention provides a process for rebalancing the electrolytes of an RFC wherein the RFC comprises a halogen/halide electrolyte in one half of the cell and a sulfide/polysulfide electrolyte in the other half of the cell, the two halves of the cell being separated by a cation-exchange membrane, and wherein the process comprises oxidising the halogen/halide electrolyte so as to convert halide to halogen and/or oxidising the sulfide/polysulfide electrolyte so as to convert sulfide to polysulfide.

The oxidation of halide to halogen rebalances the electrolytes by restoring the halogen which was reduced by reaction with migrating sulfide ions. Oxidation of halide to halogen may also be thought of as charging the halogen/halide electrolyte since the chemical content of the halogen/halide electrolyte changes in the same manner as when the RFC is in its charging cycle. The oxidation of sulfide to sulfur rebalances the electrolytes by oxidising the equivalent amount of sulfide which would ordinarily have been oxidised by the halogen which was reduced by reaction with migrating sulfide ions. Oxidation of polysulfide to sulfur may also be thought of as

discharging the sulfur/polysulfide electrolyte since the chemical content of the sulfur/polysulfide electrolyte changes in the same manner as in the RFC when it is in its discharging cycle.

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The rebalancing process may be applied continuously to the RFC wherein a sidestream of one or both electrolytes drawn from the mainstream is diverted through apparatus suitable for carrying out the 10 rebalancing process. The rebalancing process may also be applied as a batch process wherein a portion of one or both electrolytes is removed from the RFC and treated in separate apparatus suitable for carrying out the process before being returned to the RFC.

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Preferably, the oxidation of the halogen/halide and/or sulfur/polysulfide electrolyte does not result in the introduction into the electrolyte of chemical species not normally contained therein. That is to say, the 20 oxidation does not involve the addition of oxidative species or the generation of any by-products which cannot be readily removed from the electrolyte(s).

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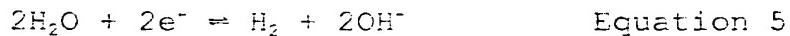
Thus, in a preferred embodiment, the electrolyte(s) is/are reacted with oxygen in the presence of a catalyst which is insoluble in the electrolyte(s). Such catalytic materials include, for example, a solid phase reactor bed through which the electrolyte(s) is/are passed together with oxygen. Examples of such 30 reactor beds which may be used to oxidise the sulfide/polysulfide electrolyte are those comprising activated carbon which selectively oxidise sulfide to sulfur. Such reactor beds have previously been used in the pulp and paper industry and are described in US-A-4024229, US-A-4073748 and US-A-1392720.

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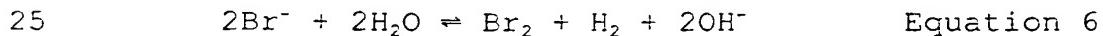
Alternatively, oxygen may be introduced to the electrolyte in the form of hydrogen peroxide, the

decomposition of which does not introduce species to the electrolyte which are not normally contained therein.

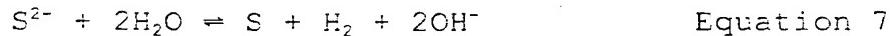
- 5 In another preferred embodiment the halogen/halide electrolyte and/or sulfide/polysulfide electrolyte is removed from the RFC to one or more external electrochemical cells wherein halide is oxidised to halogen and/or sulfide is oxidised to sulfur as a  
10 half-cell reaction in an electrochemical process. It will be understood by those skilled in the art that a number of reduction half-cell reactions may be used to counter the oxidation of the halide and/or sulfide. However, in a preferred embodiment, the other half-cell reaction involves the reduction of water to  
15 hydrogen and hydroxide ions according to the half-cell reaction shown in Equation 5 below:



- 20 Thus the preferred oxidation processes may be represented by the reactions shown in Equations 6 and 7 below:



and/or



- 30 Electrochemical oxidation is particularly preferred because no additional reagents which might contaminate the electrolytes are involved.

- 35 Preferably, the process comprises treatment of the halogen/halide electrolyte by electrochemical oxidation.

- It will be appreciated that, although the process of oxidising the halogen/halide and/or sulfide/polysulfide electrolytes can be used to rebalance the electrolytes, there is still a net loss of active sulfur species from the cell. This is because the sulfide and polysulfide ions which cross to the bromine electrolyte and are oxidised to sulfate ions are not recovered. Thus, in a preferred embodiment of the present invention, the process 10 additionally comprises adding elemental sulfur or a sulfide salt to the sulfide/polysulfide electrolyte in an amount such as to restore the initial concentration of active sulfur species.
- 15 The present invention also includes within its scope an electrochemical process for energy storage and/or power delivery comprising the steps of:
- (i) maintaining and circulating electrolyte flows in a fully liquid system in which the active constituents are fully soluble in a single cell or in an array of repeating cell structures, each cell with a chamber (+ve chamber) containing an inert +ve electrode and a chamber (-ve chamber) containing an inert -ve electrode, the chambers being separated from one another by an ion exchange membrane, the electrolyte circulating in the -ve chamber of each cell during power delivery containing a sulfide, and the electrolyte circulating in the +ve chamber during power delivery containing bromine as an oxidising agent,
- (ii) restoring or replenishing the electrolytes in the +ve and -ve chambers by circulating the electrolyte from each chamber to storage means comprising a volume of electrolyte greater than the cell volume for extended

delivery of power over a longer discharge cycle than the cell volume alone would permit, and  
(iii) rebalancing the electrolytes.

Preferably, the electrolytes are rebalanced according to the process described above.

The present invention also includes within its scope apparatus for carrying out a process as described above, the apparatus comprising:

- (i) a single cell or an array of repeating cell structures, each cell with a chamber (+ve chamber) containing an inert +ve electrode and a chamber (-ve chamber) containing an inert -ve electrode, the chambers being separated from one another by an ion exchange membrane, the electrolyte circulating in the -ve chamber of each cell during power delivery containing a sulfide, and the electrolyte circulating in the +ve chamber during power delivery containing bromine as an oxidising agent,
  - (ii) storage and circulation means for each electrolyte for restoring or replenishing the electrolytes in the +ve and -ve chambers, and  
(iii) means for rebalancing the electrolytes.
- Preferably the means for rebalancing the electrolytes comprises one or more solid phase reactor beds through which one or both electrolyte(s) is/are passed together with oxygen. More preferably, when the sulfide/polysulfide electrolyte is to be oxidised, the one or more solid phase reactor beds comprise activated carbon.

In another preferred embodiment the means for rebalancing the electrolytes comprises one or more external electrochemical cells.

5 The present invention will be further described with reference to the accompanying drawings in which:

Fig 1A is a schematic view of a basic electrochemical reduction-oxidation cell in which a sulfide/polysulfide reaction is carried out in one 10 half of the cell and a bromine/bromide reaction is carried out in the other half of the cell;

Fig 1B is a diagram of cell arrays using the system of 15 Fig 1A;

Fig 2 is a block diagram of a fluid flow system using the cell of Fig 1A;

20 Fig 3 is a flow diagram of an apparatus for carrying out a preferred embodiment of the process of the present invention.

Fig 4 is a flow diagram of an apparatus for carrying 25 out a preferred embodiment of the process of the present invention.

30 Fig 5 is a flow diagram of an apparatus for carrying out a preferred embodiment of the process of the present invention.

Fig 6 is a flow diagram of an apparatus for carrying 35 out a preferred embodiment of the process of the present invention.

Fig 7 is a graph of voltage versus time for a selected number of cycles of a RFC which does not incorporate a

rebalancing process in accordance with the present invention.

Fig 8 is a graph of voltage versus time for a selected number of cycles of a RFC which does incorporate a rebalancing process in accordance with the present invention.

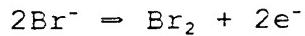
Fig 1A shows a cell 10 with a positive (+ve) electrode 12 and a negative (-ve) electrode 14 and a cation exchange membrane 16 which may be formed from a fluorocarbon polymer with sulfonic acid functional groups to provide charge carriers. The membrane 16 acts to separate the +ve and -ve sides of the cell 10 and is selected to minimize migration of bromine from the +ve side to the -ve side and to minimize migration of S<sup>2-</sup> ions from the -ve side to the +ve side. An aqueous solution 22 of NaBr is provided in a chamber 22C formed between the +ve electrode 12 and the membrane 16 and an aqueous solution 24 of Na<sub>2</sub>S<sub>x</sub> is provided in a chamber 24C formed between the -ve electrode 14 and the membrane 16. A K<sub>2</sub>S<sub>x</sub> solution, which is more soluble and more expensive than the Na<sub>2</sub>S<sub>x</sub> solutions, is used in another embodiment.

When the cell is in the discharged state, a solution of NaBr of up to 6.0 molar concentration exists in the chamber 22C of the cell and a solution of Na<sub>2</sub>S<sub>5</sub> at 0.5 to 1.5 molar, exists in chamber 24C of the cell. Higher molarity is possible with K<sub>2</sub>S<sub>5</sub>.

As the cell is charged, Na<sup>+</sup> ions are transported through the cation membrane 16, as shown in Fig 1A, from +ve to the -ve side of the cell. Free bromine is produced via oxidation of the bromide ions at the +ve electrode and dissolves as a tribromide or pentabromide ion. Sulfur is reduced at the -ve

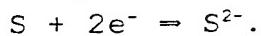
electrode and the pentasulfide,  $\text{Na}_2\text{S}_5$ , salt eventually becomes the monosulfide as the charging proceeds to completion. At the +ve side the following reaction occurs,

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and at the -ve side the following reaction occurs,

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The membrane separates the two electrolytes and prevents bulk mixing and also retards the migration of sulfide ions from the -ve side to the +ve side, and the migration of  $\text{Br}^-$  and  $\text{Br}_2$  from the +ve to the -ve side. Diffusion of the sulfide ions results in coulombic loss and results in the oxidation of some of the sulfide content of the system to sulfate ions. It is this oxidation which results in the electrolytes becoming unbalanced.

20

When providing power, the cell is discharging. During this action, reversible reactions occur at the two electrodes. At the +ve side electrode 12, bromine is reduced to  $\text{Br}^-$ , and at the -ve electrode, the  $\text{S}^{2-}$  ion is oxidized to molecular S. The electrons produced at the -ve electrode form the current through a load. The chemical reaction at the +ve electrode produces 1.06 to 1.09 volts and the chemical reaction at the -ve electrode produces 0.48 to 0.52 volts. The combined chemical reactions produce an open circuit voltage of 1.54 to 1.61 volts per cell.

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The energy density of the bromine/sulfur couple will be limited by the permissible maximum concentration of the  $\text{Br}_2$  in the +ve side, not by the solubilities of the constituent salts, such as  $\text{NaBr}$  and  $\text{Na}_2\text{S}$ , which are

high.

The reacting ions are  $S^{2-}$  and  $Br^-$  going back and forth to the elemental stage during the oxidation/reduction processes. The cation which is associated with them essentially takes no part in the energy producing process. Hence, a cation of "convenience" is chosen. Sodium or potassium are preferred choices. Sodium and potassium compounds are plentiful, they are inexpensive and have high water solubilities. Lithium and ammonium salts are also possibilities, but at higher costs.

Fig 1B shows an array 20 of multiple cells connected in electrical series and fluid parallel. Multiple mid-electrodes 13 (each one having a +ve electrode side 12A and -ve electrode side 14A) and end electrodes 12E (+ve) and 14E (-ve) are spaced out from each other by membranes 16 and screen or mesh spacers (22D, 24D) in all the cell chambers 22C, 24C, (portions of two of which 22D, 24D are shown by way of example) to form end cells  $C_{E1}$  and  $C_{E2}$  and an array of mid cells  $C_M$  (typically 10-20; but note much smaller and much higher numbers of cells can be accommodated). The end electrodes 12E (+ve) and 14E (-ve) have internal conductors 12F and 14F (typically copper screens) encapsulated therein and leading to external terminals 12G, 14G which are connected to external loads (e.g. to motor(s) via a control circuit (CONT), the motor(s) may be used to drive a vehicle) or power sources (e.g. utility power grid when used as a load-levelling device).

Fig 2 shows a free flow system, a power generation/storage system utilizing one or more of the batteries or cell array formats 20. Each cell 20 receives electrolyte through pumps 26 and 28 for the

NaBr and Na<sub>2</sub>S<sub>2</sub> solutions (22 and 24, respectively). The electrolytes 22 and 24 are stored in containers 32 and 34. The tanks 32, 34 can be replaced with freshly charged electrolyte by substituting tanks containing fresh electrolyte and/or refilling them from charged supply sources via lines 32R, 34R with corresponding lines (not shown) provided for draining spent (discharged) reagent. The electrolytes 22 and 24 are pumped from tanks 32 and 34, respectively, into the 10 respective chambers 22C and 24C by means of pumps 26 and 28.

Fig 3 shows a free flow system in which an array of 15 cells 20 are supplied with halogen/halide and sulfide/polysulfide electrolyte from storage tanks 41 and 42 via lines 43 and 44. Sulfide/polysulfide electrolyte may be removed from storage tank 42 via line 45 which transfers it to a catalyst reactor bed 46. Air is also introduced into the catalyst bed and 20 the oxygen contained in the air, in conjunction with the catalyst in the bed causes oxidation of the sulfide/polysulfide electrolyte as it passes through the bed. A line 47 transports oxidised sulfide/polysulfide electrolyte from the catalyst bed 25 and returns it to the storage tank 42.

Fig 4 shows a free flow system in which an array of 30 cells 20 are supplied with halogen/halide and sulfide/polysulfide electrolyte from storage tanks 41 and 42 via lines 43 and 44. Halogen/halide electrolyte may be removed from storage tank 41 via line 50 which transfers it to an external electrochemical cell 51 wherein the halide is oxidised to halogen as a half-cell reaction in an electrochemical process. The other 35 half-cell reaction involves the reduction of water to hydrogen and hydroxide ions. An aqueous electrolyte is stored in tank 52 and transported to the

electrochemical cell 51 via line 53. The reduced electrolyte is passed via line 54 to tank 55 where hydrogen gas which is generated by the electrochemical reaction may be vented from the system. The 5 electrolyte returns via line 56 to storage tank 52.

Fig 5 shows a particularly preferred variation of the free flow system illustrated in Fig 4. In this embodiment the aqueous electrolyte which is reduced in 10 the external electrochemical cell 51 is also halogen/halide electrolyte which has been removed from storage tank 41 to tank 52 via line 57. In this case, the reduction reaction will initially involve reduction of any residual halogen to halide and will 15 subsequently involve reduction of water to hydrogen and hydroxide ions.

Fig 6 shows another preferred variation of the free flow system and is essentially a combination of the systems illustrated in Figs 4 and 5. In this embodiment the halogen/halide electrolyte is passed through two external electrochemical cells 51 and 51'. The electrolyte which is reduced in the first external cell 51 is supplied from the halide/halogen storage 25 tank 41 and the electrolyte which is reduced in the second external cell 51' is an aqueous solution stored in tank 52'.

The present invention will now be further described by 30 reference to the following examples.

Comparative Example 1

A regenerative fuel cell of the type described above having sulfide/polysulfide and bromine/bromide 35 electrolytes was set up. The cell had the following specifications:

electrode material: polyethylene impregnated with  
activated carbon

electrode area: 2000cm<sup>2</sup>

current density: 80mA/cm<sup>2</sup>

5       electrolyte volume: 91 per electrolyte

cycle time: 6hours (i.e. 3hours charge  
and 3hours discharge)

flow rate: 1000ml/min

membrane material: Nafion 115™

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The cell was operated over 18 cycles (108 hours) and  
the cell voltage was monitored throughout this period.

The results are shown in Fig.7. It can be seen that  
after a limited number of cycles (about 7) the cell  
voltage limits early on the discharge cycle due to the  
lack of bromine. This problem gets worse as the  
number of cycles increases. It can be clearly seen on  
the graph that after about 12 cycles (72 hours) the  
cell fails to maintain a good voltage performance over  
the whole of the 3hour discharge cycle.

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#### Example 1

An identical RFC to that used in Comparative Example 1  
was set up. This time the electrolytes were

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continuously rebalanced by oxidation of a sidestream  
of the bromide/bromine electrolyte drawn from the  
mainstream. Oxidation occurred in one half of an  
external electrochemical cell wherein the electrolyte  
undergoing reduction in the other half of the cell was  
dilute aqueous sodium hydroxide. The external  
30       electrochemical cell used was an MP cell from  
Electrocell AB having the following specifications:

35       anode material: platinum

cathode material: nickel

electrode area: 100cm<sup>2</sup>

current density: 13mA/cm<sup>2</sup>

flow rate: 270ml/min  
membrane material: Nafion 350™

Fig.4 shows a schematic representation of the  
5 apparatus used in the present example. The cell was  
operated over at least 92 cycles (552 hours) and the  
cell voltage was monitored throughout this period. The  
results from the period from 400 to 550 hours are  
shown in Fig.8. It can be seen that even after 91  
10 cycles (546 hours) the cell voltage does not limit  
early on the discharge cycle as occurred in the  
unbalanced cell. The cell retains a good voltage  
performance over the whole of the 3hour discharge  
cycle.

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Claims

1. A process for rebalancing the electrolytes of a regenerative fuel cell (RFC) wherein the RFC comprises a halogen/halide electrolyte in one half of the cell and a sulfide/polysulfide electrolyte in the other half of the cell, the two halves of the cell being separated by a cation-exchange membrane, and wherein the process comprises oxidising the halogen/halide electrolyte so as to convert halide to halogen and/or oxidising the sulfide/polysulfide electrolyte so as to convert sulfide to polysulfide.
- 15 2. A process as claimed in claim 1 wherein oxidation of the halogen/halide and/or sulfur/polysulfide electrolyte is carried out on a portion of the electrolyte which has been removed from the RFC either continuously or in a batch process and which is subsequently returned to the RFC.
- 20 3. A process as claimed in claim 1 or claim 2 wherein oxidation of the halogen/halide and/or sulfur/polysulfide electrolyte does not result in the introduction into the electrolyte(s) of chemical species which cannot be readily removed from the electrolyte(s).
- 25 4. A process as claimed in claim 3 wherein the electrolyte(s) is/are reacted with oxygen in the presence of a catalyst which is insoluble in the electrolyte(s) and which does not generate by-products which are soluble in the electrolyte(s).
- 30 5. A process as claimed in claim 4 wherein the electrolyte(s) is/are reacted in the presence of

a solid phase reactor bed through which the electrolyte(s) is/are passed together with oxygen.

- 5        6. A process as claimed in claim 5 wherein the reactor bed comprises activated carbon.
- 10      7. A process as claimed in claim 3 wherein the electrolyte(s) is/are reacted with hydrogen peroxide.
- 15      8. A process as claimed in any one of claims 1 to 3 wherein the halogen/halide electrolyte and/or sulfide/polysulfide electrolyte is removed from the RFC to one or more external electrochemical cells wherein halide is oxidised to halogen and/or sulfide is oxidised to polysulfide as a half-cell oxidation reaction in an electrochemical process.
- 20      9. A process as claimed in claim 8 wherein the half-cell reduction reaction of the electrochemical process is the reduction of water to hydrogen and hydroxide.
- 25      10. A process as claimed in any one of the preceding claims which additionally comprises adding elemental sulfur and/or a sulfide salt to the sulfide/polysulfide electrolyte in an amount sufficient to restore the initial concentration of sulfur species.
- 30      11. An electrochemical process for energy storage and/or power delivery comprising the steps of:  
35            (i) maintaining and circulating electrolyte flows in a fully liquid system in which the active constituents are fully soluble in a

- single cell or in an array of repeating cell structures, each cell with a chamber (+ve chamber) containing an inert +ve electrode and a chamber (-ve chamber) containing an inert -ve electrode, the chambers being separated from one another by an ion exchange membrane, the electrolyte circulating in the -ve chamber of each cell during power delivery containing a sulfide, and the electrolyte circulating in the +ve chamber during power delivery containing bromine as an oxidising agent,
- (ii) restoring or replenishing the electrolytes in the +ve and -ve chambers by circulating the electrolyte from each chamber to storage means comprising a volume of electrolyte greater than the cell volume for extended delivery of power over a longer discharge cycle than the cell volume alone would permit, and
- (iii) rebalancing the electrolytes.
12. A process as claimed in claim 11 wherein the electrolytes are rebalanced according to a process as claimed in any one of claims 1 to 10.
13. Apparatus for carrying out a process as claimed in claim 11 or 12 comprising:
- (i) a single cell or an array of repeating cell structures, each cell with a chamber (+ve chamber) containing an inert +ve electrode and a chamber (-ve chamber) containing an inert -ve electrode, the chambers being separated from one another by an ion exchange membrane, the electrolyte circulating in the -ve chamber of each cell during power delivery containing a sulfide,

and the electrolyte circulating in the +ve chamber during power delivery containing bromine as an oxidising agent.

5 (ii) storage and circulation means for each electrolyte for restoring or replenishing the electrolytes in the +ve and -ve chambers, and

(iii) means for rebalancing the electrolytes..

10 14. Apparatus as claimed in claim 13 wherein the means for rebalancing the electrolytes comprises one or more solid phase reactor beds through which one or both electrolyte(s) is/are passed together with oxygen.

15 15. Apparatus as claimed in claim 14 wherein the one or more solid phase reactor beds comprise activated carbon.

20 16. Apparatus as claimed in claim 13 wherein the means for rebalancing the electrolytes comprises one or more external electrochemical cells.

25 17. A process substantially as hereinbefore described with reference to example 1.

18. A process substantially as hereinbefore described with reference to any one of figures 3 to 6.

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FIG. 1A.

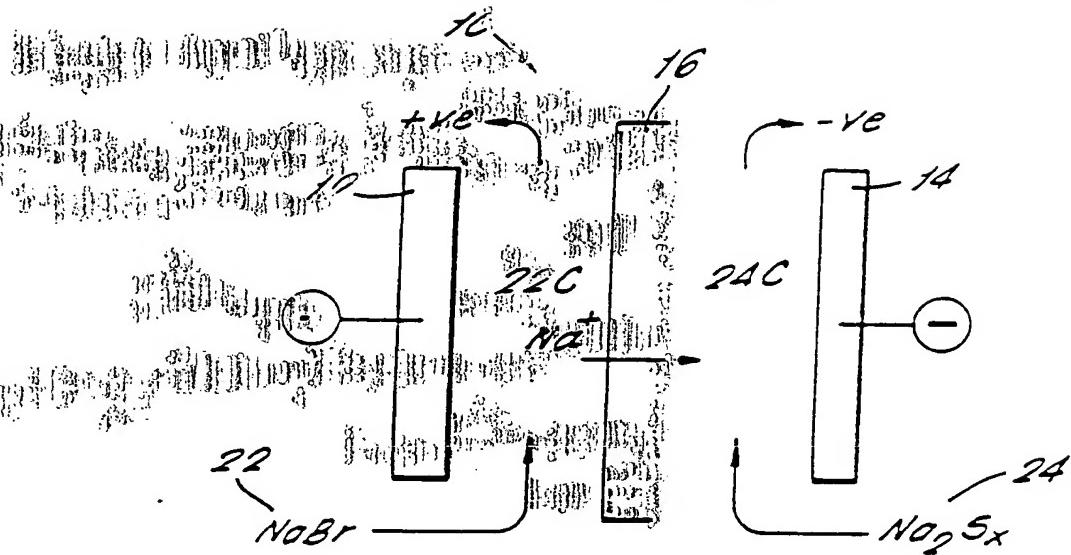
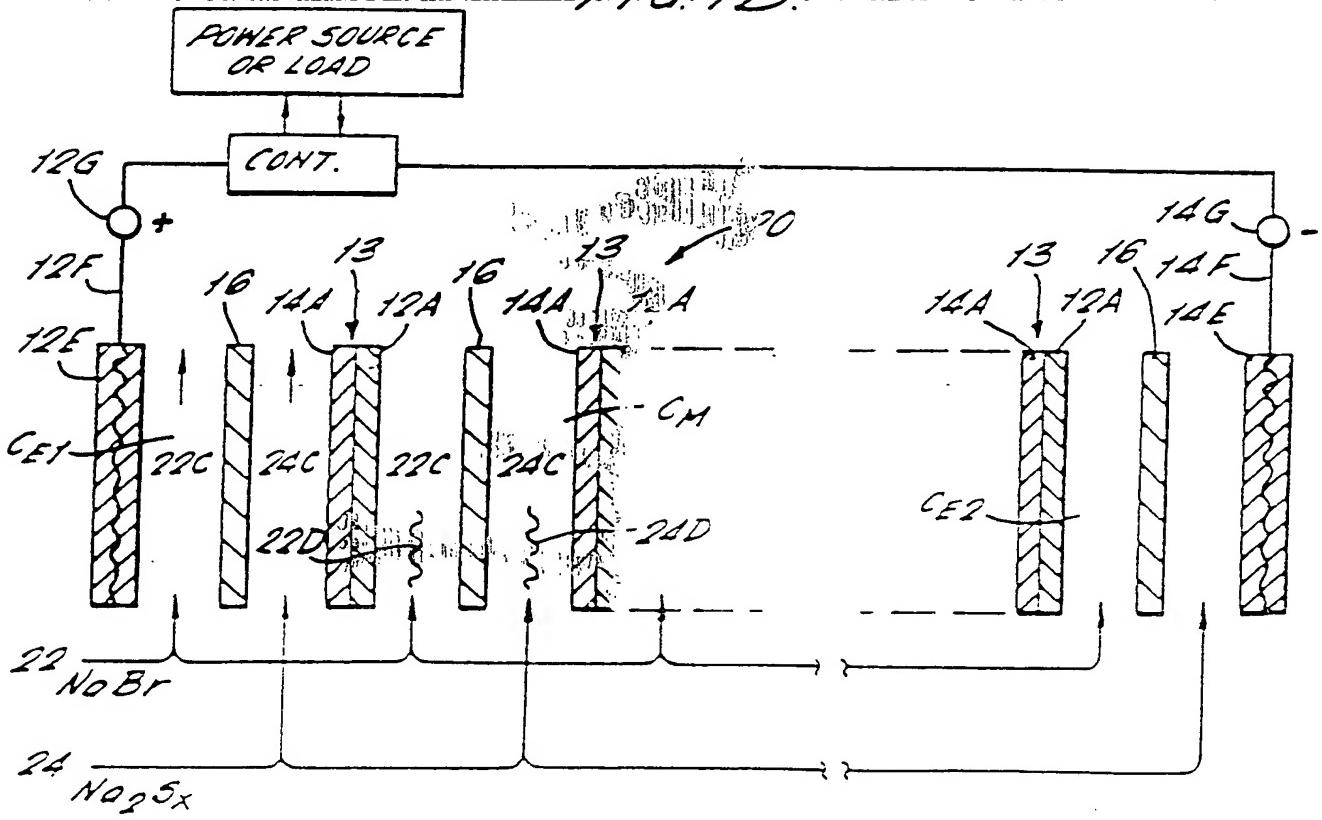


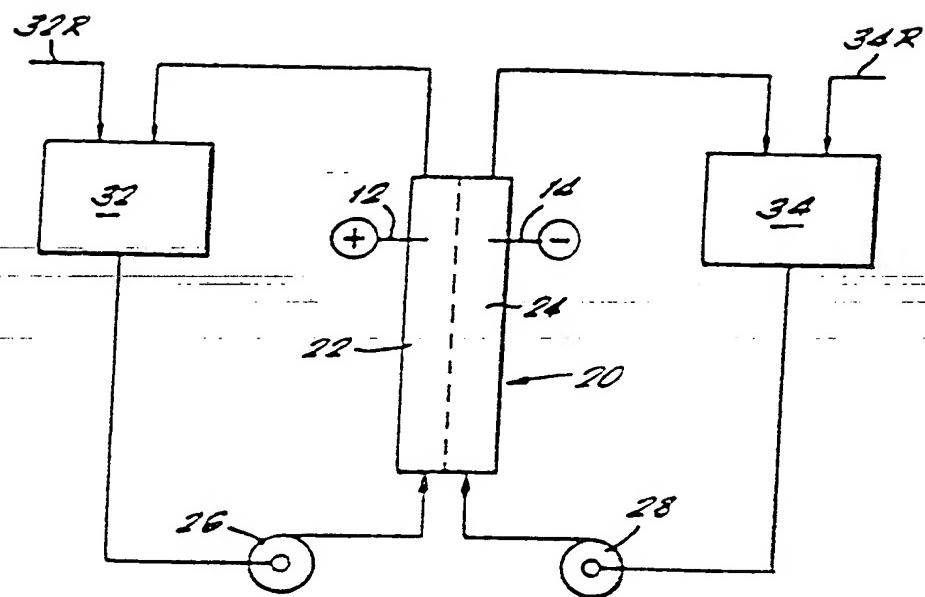
FIG. 1B.



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FIG. 2.



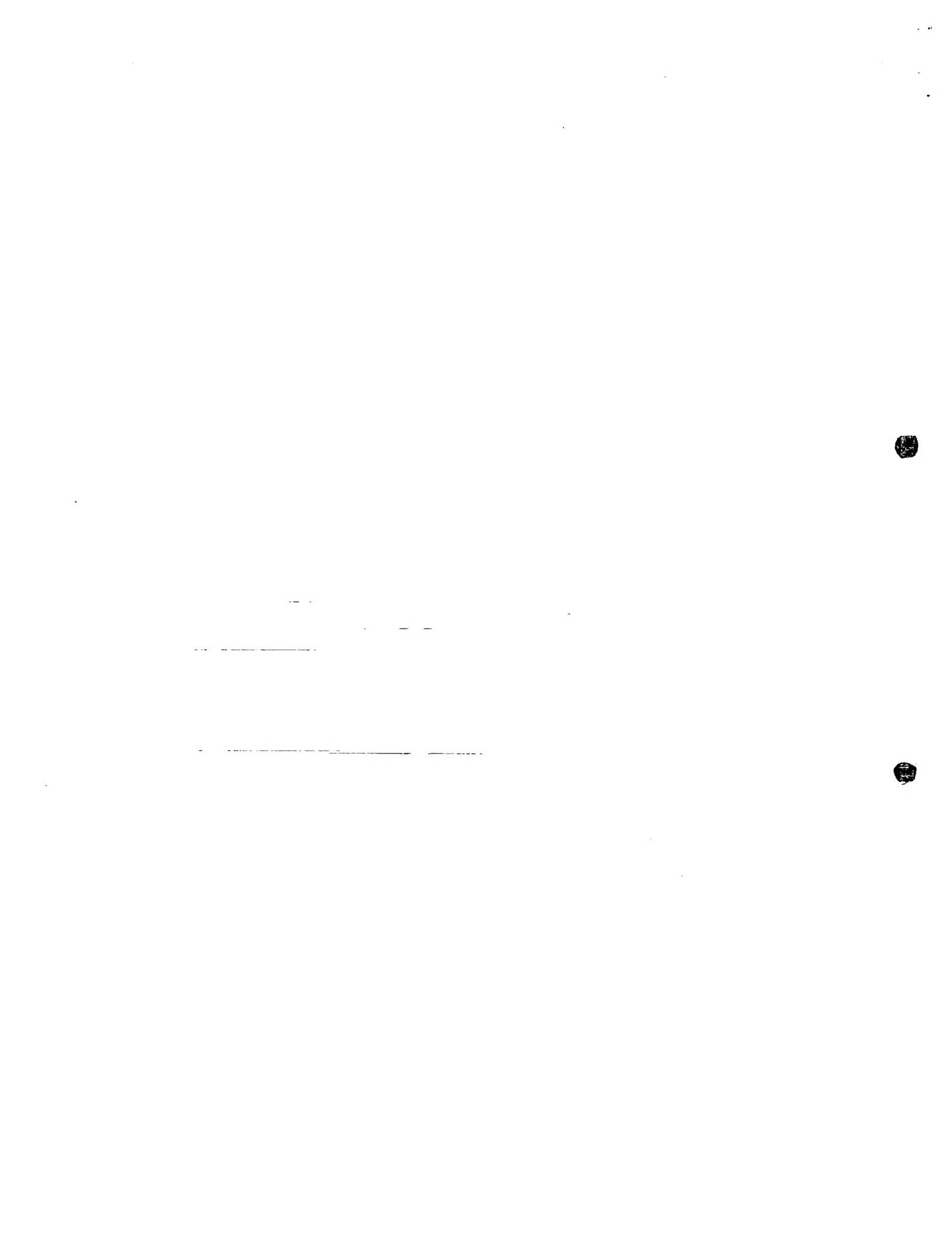
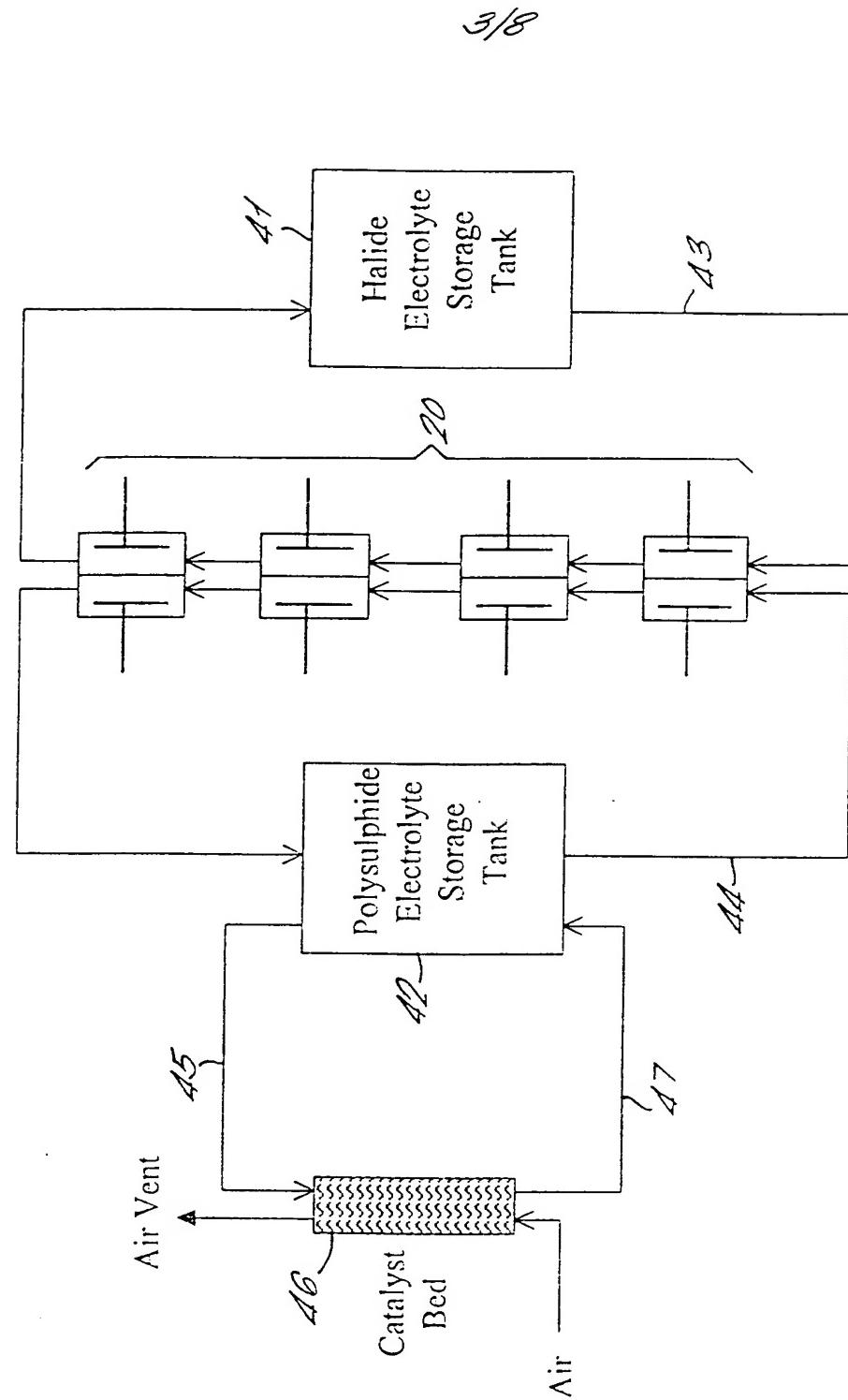


FIG. 3.



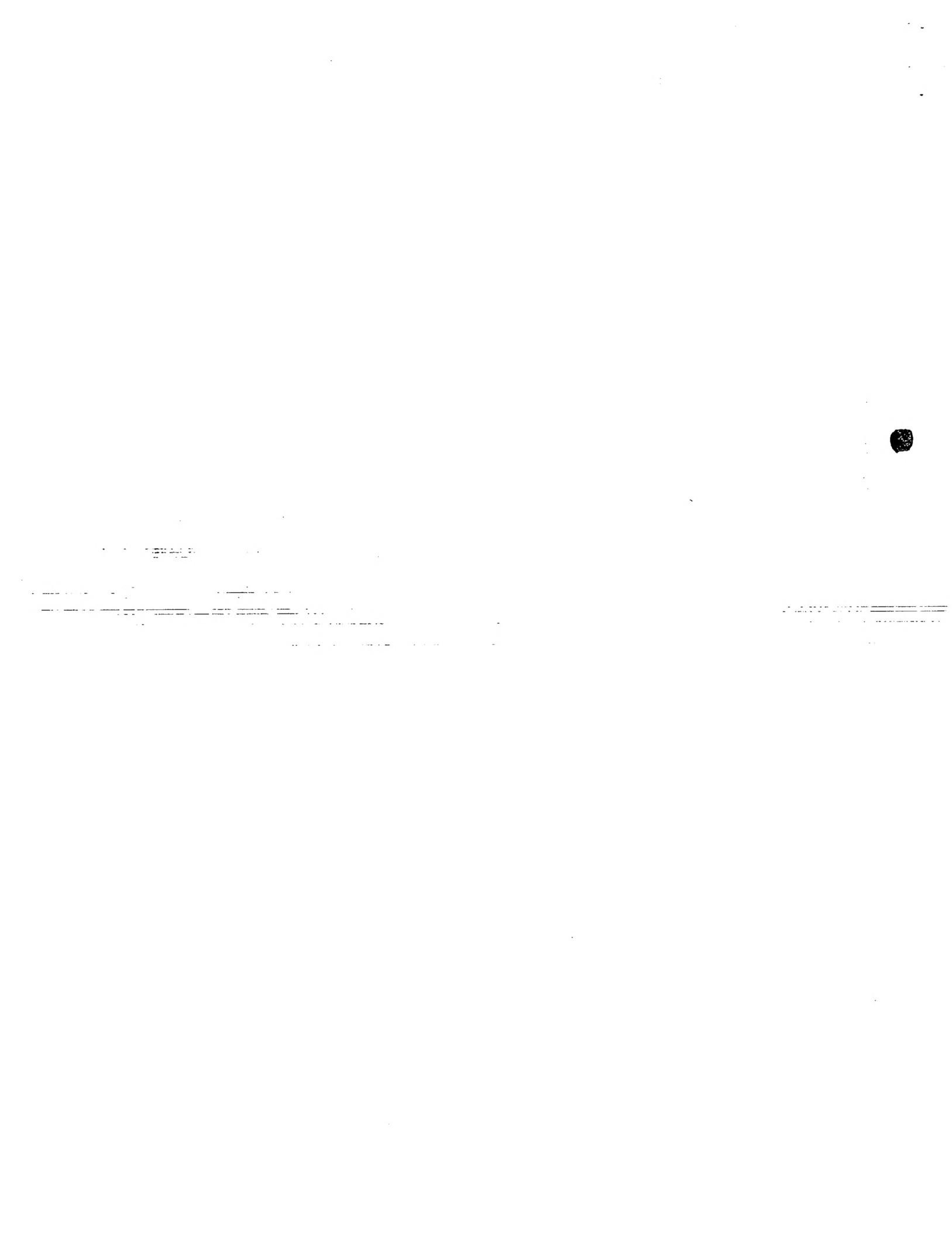
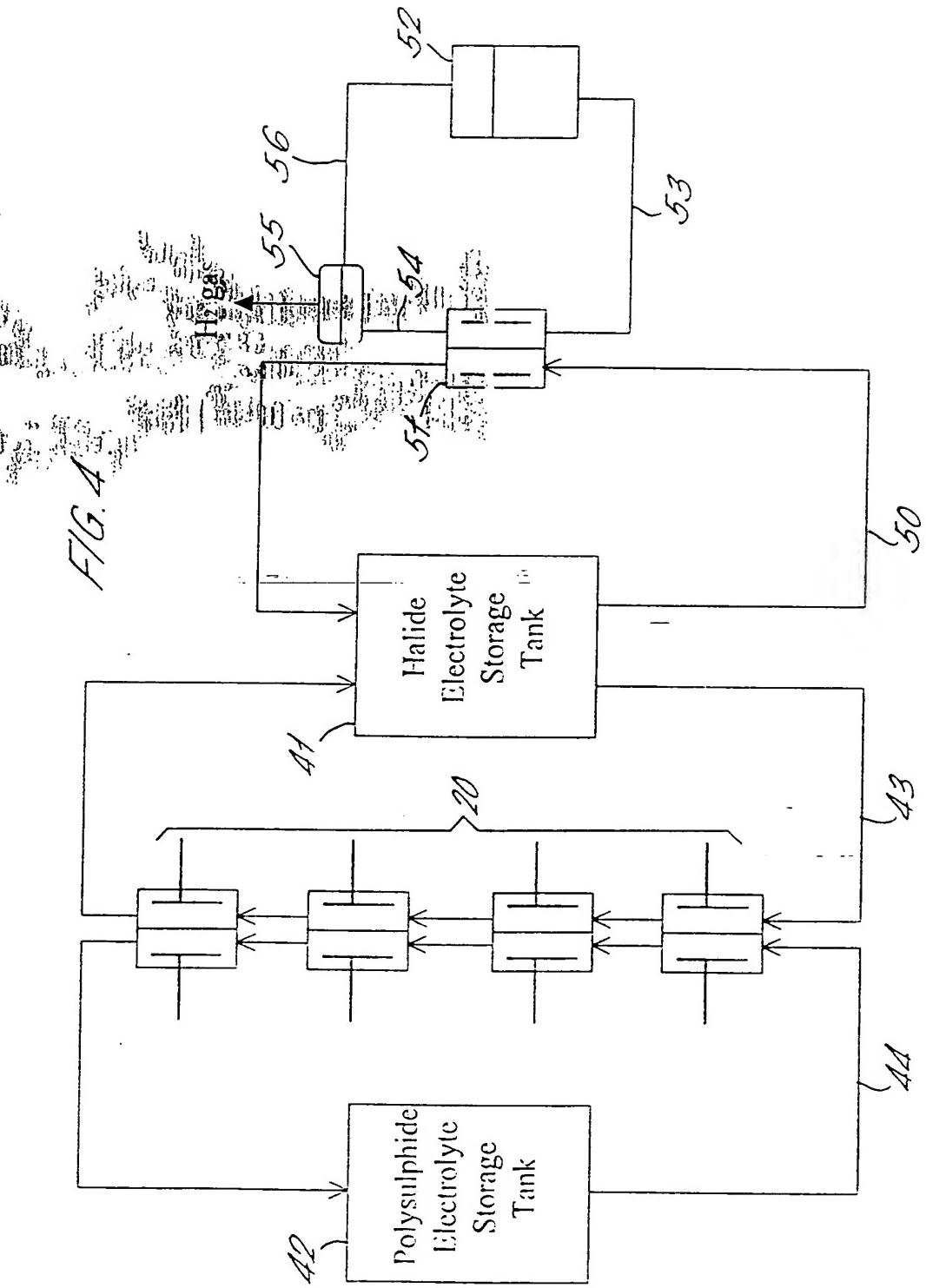


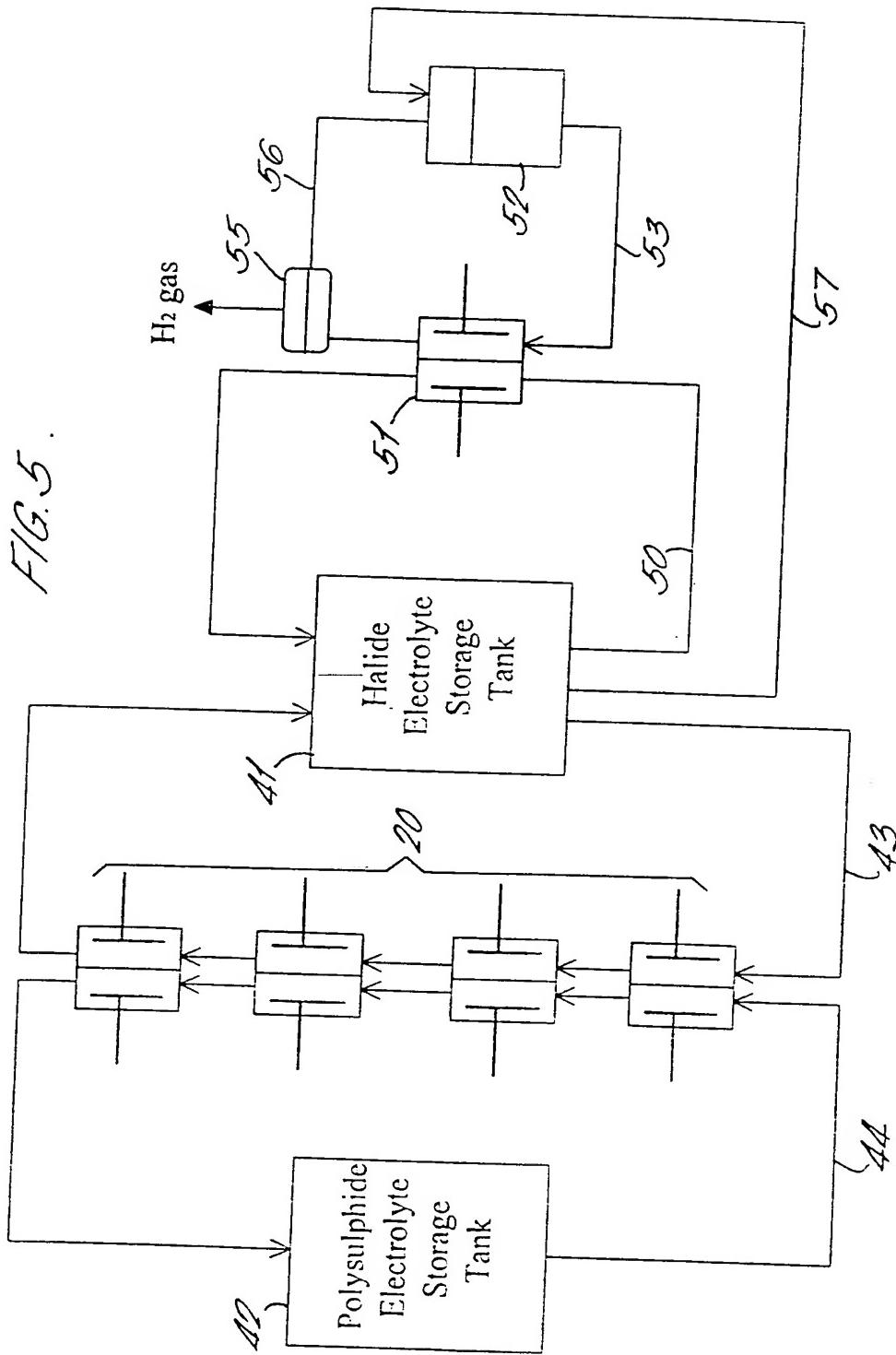
FIG. 4





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FIG. 5.



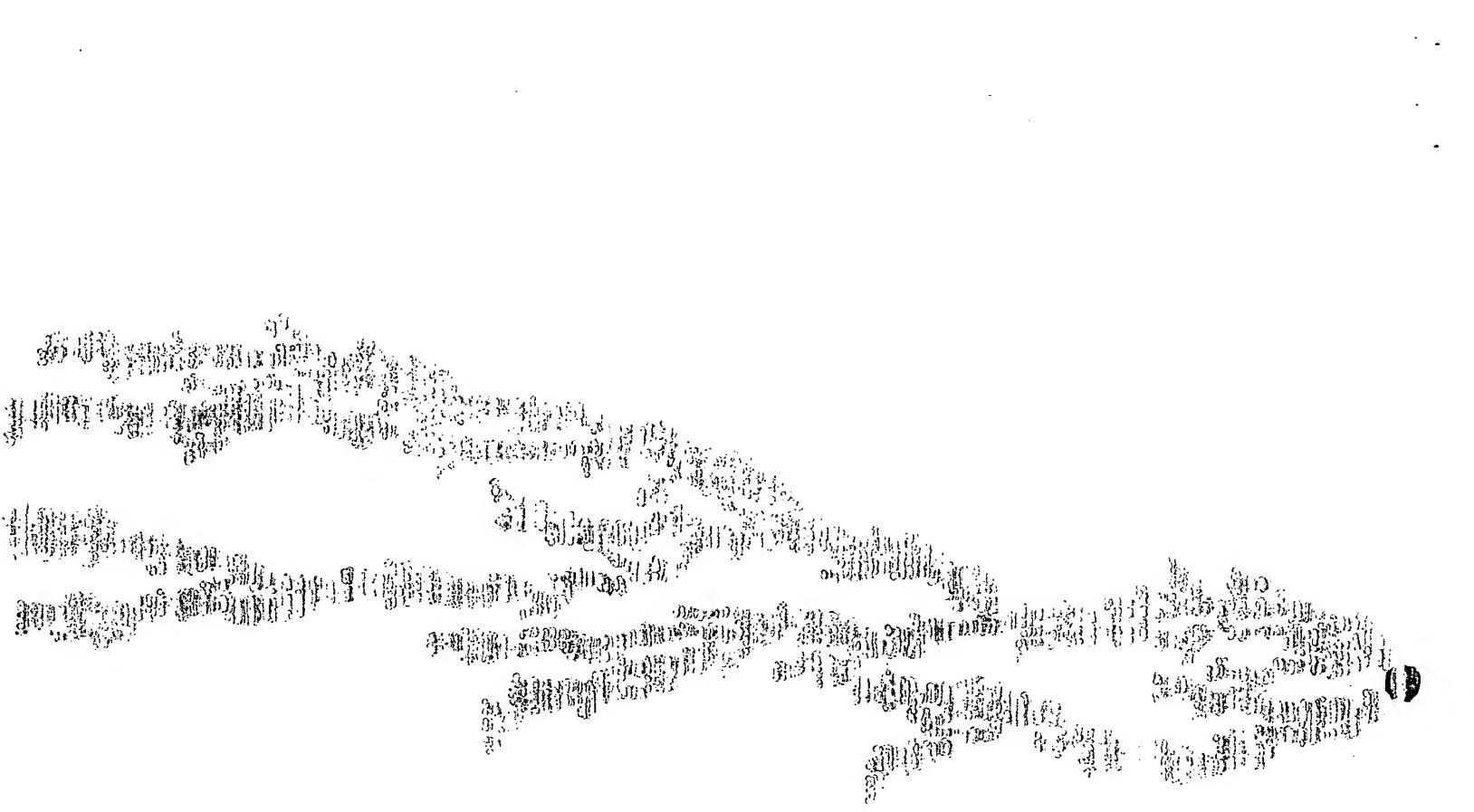
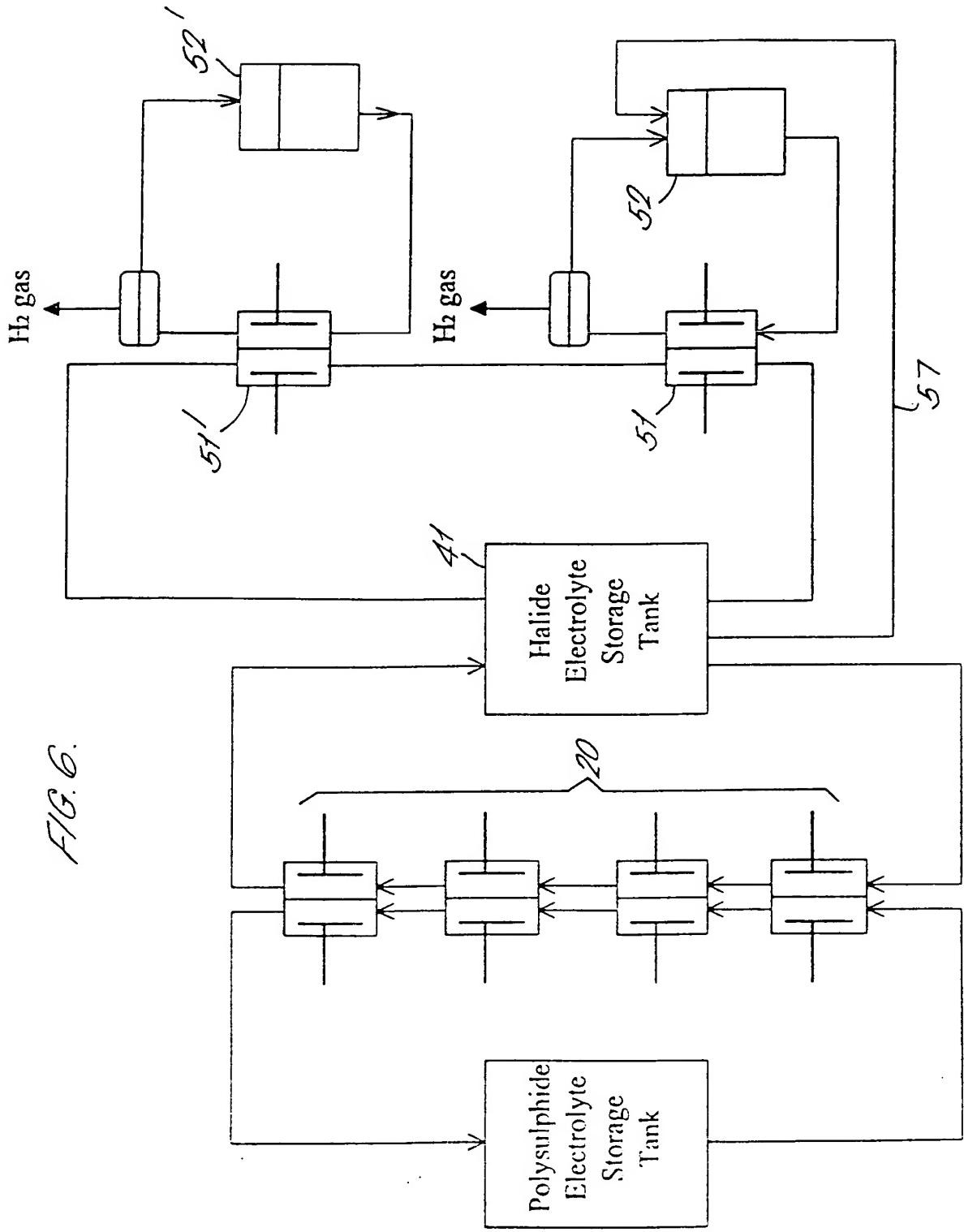


FIG. 6.



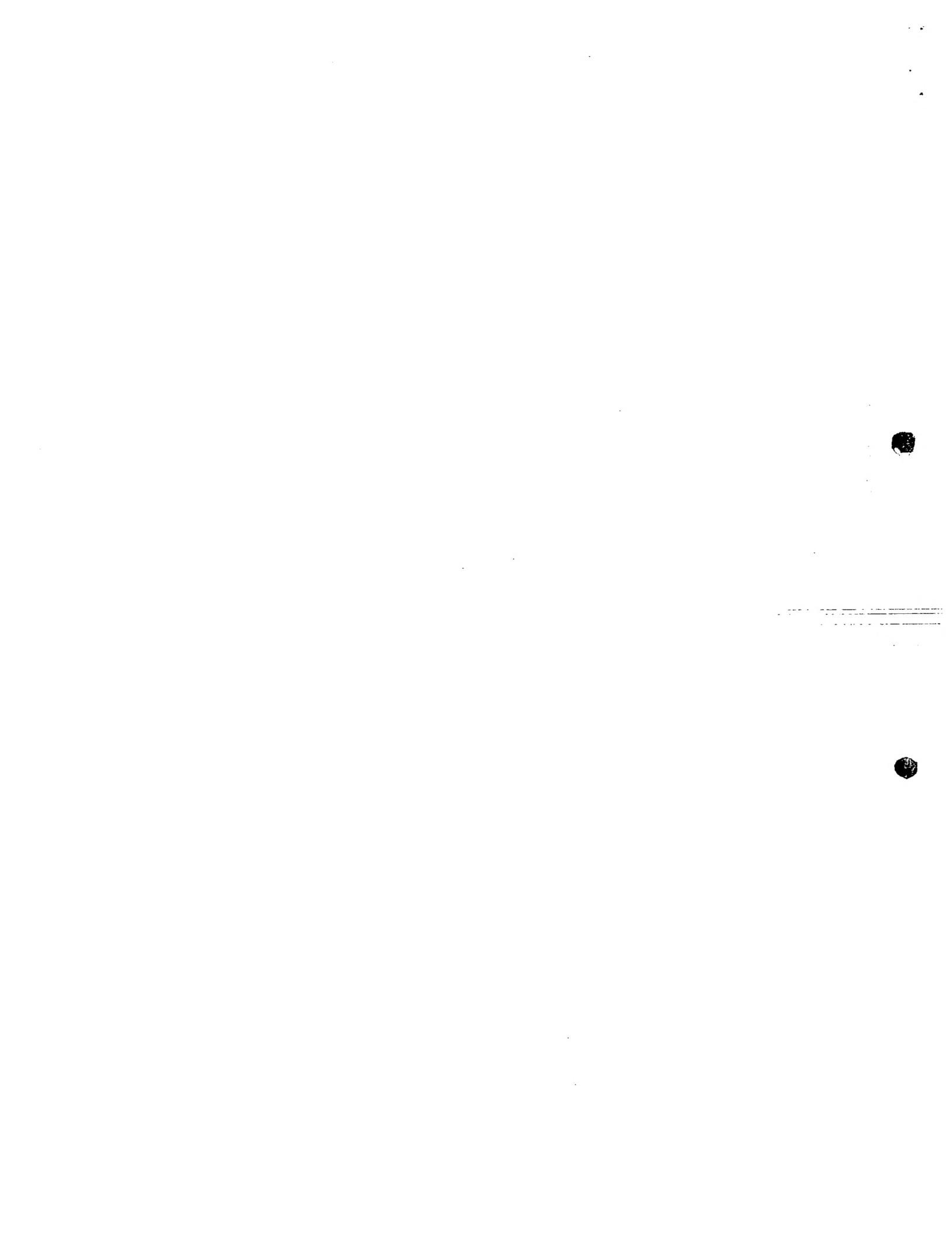


FIG. 7

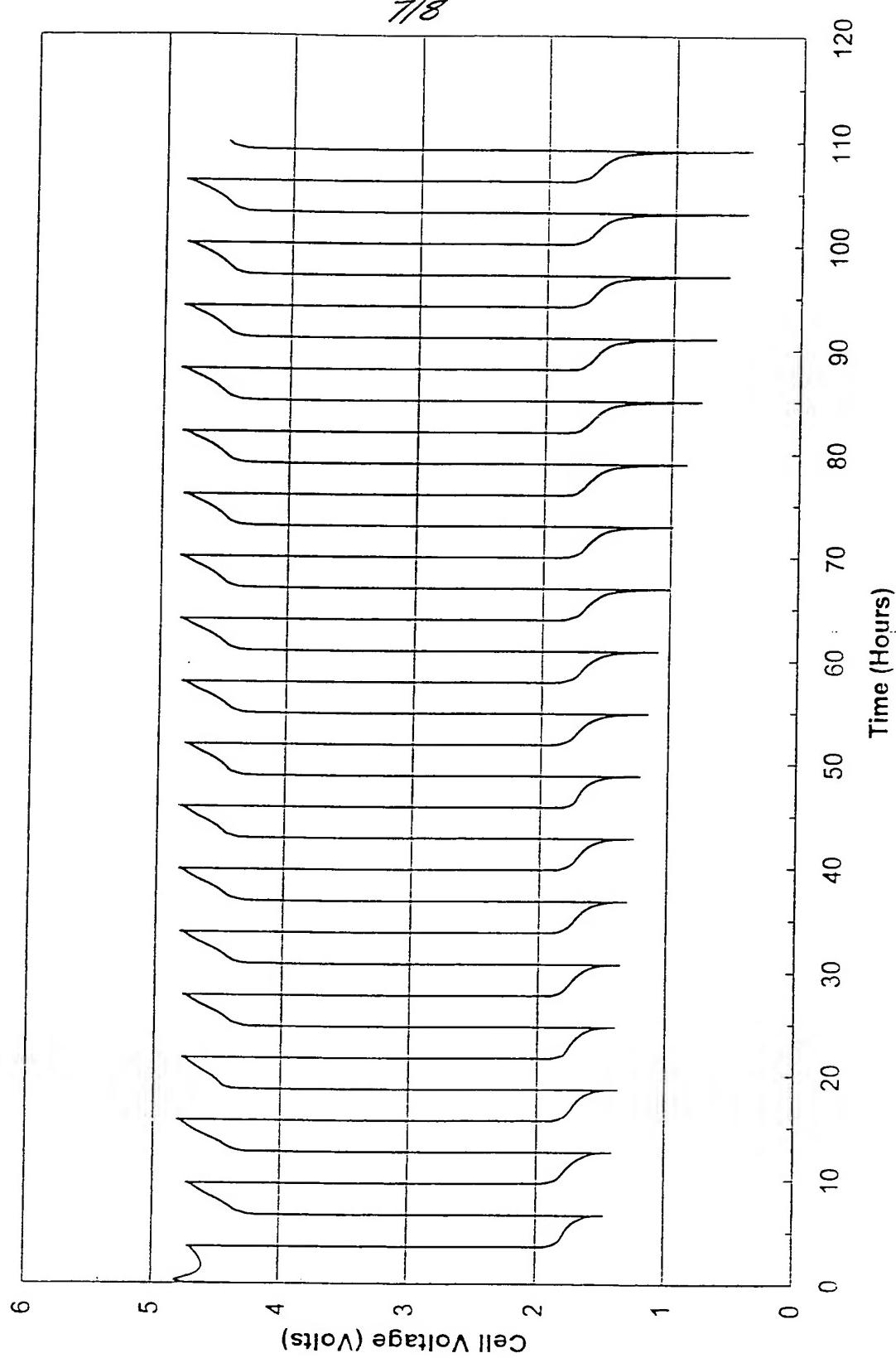




Fig. 8.

